Determination of Marine Aerosol Properties Using a Bistatic Nephelometer

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LONG-TERM GOAL

Our long-term goal is to develop methods to characterize remotely the optical properties of aerosols in the marine atmospheric boundary layer (MABL). The method is based on instruments and analytic methods that use angle-dependent polarization information from the light scattered by aerosols. The information derived from these measurements will enable accurate prediction of the aerosol optical properties and consequently their effect on light propagation in the MABL.

OBJECTIVES

The objective of this work is to develop and deploy a new light scattering instrument to remotely characterize atmospheric aerosols. The bi-static nephelometer (an instrument with separately pointed light source and detector that probes at a distance) is an effective tool to obtain data on aerosol content and light propagation in the MABL. It provides a means to probe the area about its mounting platform whether it is mounted on-shore or shipboard. Polarized light scattering techniques have the advantage of providing a direct, rapid *in situ* measurement of the optical properties of aerosols. By using various scanning strategies, the aerosols in the MABL may be characterized for visibility and the aerosol content in spatial detail (mapped) with time.

An analytical modeling effort is required to interpret the bistatic nephelometer measurements. The results of modeled calculations are used to match the scattering measurements to derive information about the atmospheric aerosols. Scattering models based on Mie calculations must be supplemented with more general analytical approaches that provide for scattering from non-spherical particles. Therefore viable models are being developed to calculate scattering from a range of regular and irregular non-spherical particles.

APPROACH

The bistatic nephelometer uses polarization modulation and synchronous detection to measure the elements of the Mueller scattering matrix for the atmospheric aerosols. The existing instrument [designed for use on sea ice] is being modified for atmospheric aerosol measurements. We have tested its operation under controlled conditions in preparation for field measurements. The polarization modulated laser light source and detector assembly are directed to a region of space or scanned systematically about the instrument platform (*eg*. ship bow and stern or two locations on shore). The technique provides direct, rapid *in situ* measurement of aerosol properties in the lower MABL. A method of intensity normalization using the time varying signals arising from polarization modulation

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provides data that enable determination of size distributions and optical properties independent of aerosol density and details of the scattering volume. By scaning the light source and detector independently, the spatial and temporal character (4D) of aerosols in the MABL about the platform may be mapped. This information can be used to determine the most effective polarimetric strategy to improve visibility and target discrimination.

Measurements of scattering in the MABL, particularly in the coastal zone, require data analysis methods to retrieve size distributions and optical properties of the scatterers. Mie scattering calculations are used with fitting algorithms for particles exhibiting spherical symmetry. To predict scattering from non-spherical particles a computer code was developed using a coupled-dipole approximation (CDA). The approach allows the calculation of scattering from regular and irregular non-spherical shapes. Particle simulation requires building particles from a multiplicity of dipoles and then calculating all their interactions.

WORK COMPLETED

The bi-static nephelometer was modified from the existing ice nephelometer developed for sea ice measurements[1,2]. It is shown schematically in Fig. 1. The laser beam first passes through a photo elastic modulator (PEM) that varies the polarization at 50 kHz. The probing laser beam is rotated in a horizontal plane by a special biprism "mirror" (not shown in the figure) designed to eliminate undesirable effects of polarization due to reflection by a traditional mirror. A computer controlled stepper motor attached to the biprism mirror varies the angle of the beam as it exits the instrument and enters the atmosphere. A computer controlled eight-inch diameter F/10 Schmidt-Cassegrain telescope collects the scattered light. The collected light from the telescope passes through polarization and laser band-pass filters before detection by a photomultiplier tube (PMT).

The ac components of the received signal from the PMT that arise from the polarization modulation are related to the Mueller matrix elements. They are synchronously detected using a lock-in amplifier. By choosing the proper combination of (1) initial polarization state and PEM position; (2) final polarizing filters; and (3) lock-in amplifier frequency, measurements can be made of all 16 elements of the Mueller matrix. For atmospheric aerosols[3,4] the measurements are limited to S_{11} (total intensity) ($\mathbf{S}_{12}$ unpolarized to linear polarization) $\mathbf{S}_{34}$ (linear to circular polarization) and $\mathbf{S}_{22}$ (linear cross polarization). A lap-top computer controls the operation of entire instrument and records the output of the lock-in amplifier and signal normalization information. The PMT can be operated at either a constant dc voltage or constant dc current mode. The constant current mode provides automatic normalization of the ac signals from the lock-in amplifier that are proportional to the desired angle dependent Mueller matrix elements.

The operation of the nephelometer was verified by laboratory measurements of well-characterized scattering systems. The measurements were performed in a water tank containing an aqueous suspension of latex spheres of known diameter. There was generally good agreement between the measured matrix elements and the model calculations. These measurements are being extended using horizontal scans in air with laboratory generated aerosols..

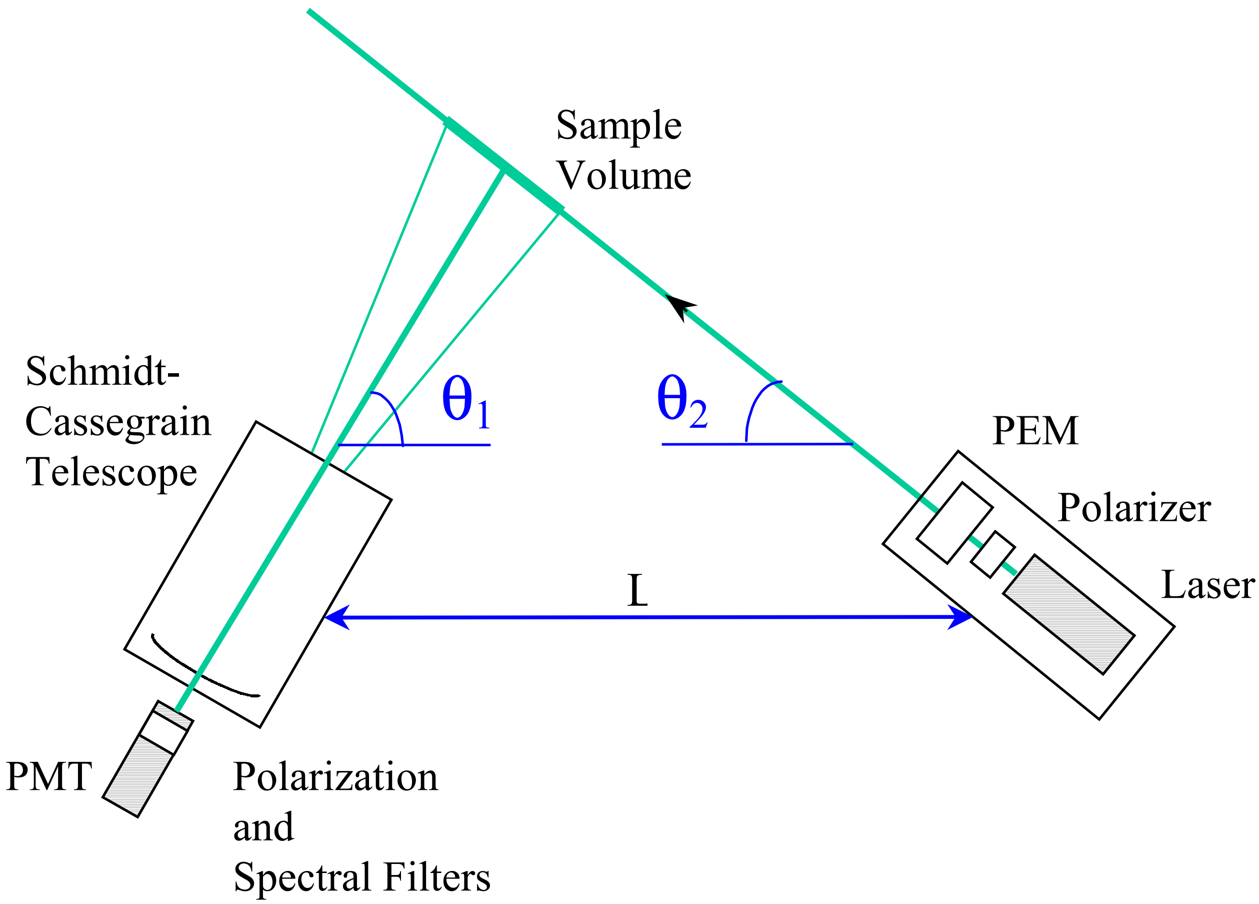

Figure 1. Bistatic Nephelometer as modified, based on original instrument developed by Miller et al. [Ref. 2]. L is the base line distance, the scattering angle is 180 – θ_1 – θ_2.

The size, refractive index and shape information for a distribution of particles from the observed aerosol scattering cannot in general be obtained by direct inversion of the Mueller matrix. It is necessary, therefore, to calculate the scattering from assumed ensembles of particles from models based on the scattering system and compare the results with the experimental data. If the particles are spherical or nearly so, then the scattering of such an ensemble can be calculated rigorously using Mie scattering models[3]. Particles may be considered spherical when $\boldsymbol{S}_{22}$ (determined by field measurement) is greater than 0.9 at all angles[5]. If the particles are spherical according to the S_{22} test, agreement between observation and calculations based on the Mie model has been found to be excellent[5]. It is anticipated in many cases that the high humidity and the proximity of the ocean water in the MABL will result in spherically symmetric particles. The measured scattering is compared with that predicted by Mie calculations and the input parameters to the model are adjusted until the agreement is optimized. The fitting procedure employed fits the three most relevant matrix elements simultaneously. The fitting is performed iteratively with a computer code developed at LBNL based on the Levenburg-Marquardt optimization technique[7]. The code uses the form of the size distribution, mean, minimum, and maximum radii and the complex refractive index as input parameters and iterates to the optimal solution based on the minimization of χ^2, the "goodness of fit" parameter. These fitting programs run rapidly on modern PCs and can done at a near real-time basis.

Theoretical analysis of the more general case of non-spherical particles is more difficult but was considered important in certain cases. MABL aerosol particles under conditions of low humidity, near shore, or in combustion-generated smoke may deviate more significantly from spherical symmetry especially as measured in fractions of the wavelength of light. In cases when the $\mathbf{S}_{22}$ measurement indicates non-spherical particles an alternative approach to modeling light scattering from non-spherical particles was developed. In this case particles are represented as an agglomeration of primary nanoparticles characterized by fractal dimension and radius of gyration. Once a particle is generated, the scattering is calculated from an ensemble average of these particles. The calculation is in this case is performed using the coupled-dipole approximation[6] (CDA) an approximation well suited for the agglomerate particle model. The calculation is more computationally intensive and therefore we used a larger machine. The calculations are carried out in parallel mode with a Cray T3E (with up to 512 processors available). The parallel codes permit simultaneous calculation of scattering from particles having different sizes, orientations, and shapes. In this case the particle size is determined using the Levenburg-Marquardt optimization technique with look-up tables for the results. Neural net calculations are being explored to obtain "first guess" information to provide more rapid calculations.

A coordinated plan for instrumental comparison and modeling is being developed for a site selected on Oahu, Hawaii in conjunction with the Universities of Hawaii and Oregon. The bistatic nephelometer will be tested and operated side by side with an imaging LIDAR and other aerosol instruments.

RESULTS

The bistatic polarization nephelometer was modified for the measurement of atmospheric aerosols in the marine atmospheric boundary layer. The instrument was tested with a water suspension of latex spheres and gave reasonable agreement with theory and further work on verifying its operation with laboratory-generated aerosols are in progress. The modeling routines being incorporated into the instrument will be ready for field trials in the near future. Calculations using the CDA adapted to parallel operation show promise in predicting and modeling the scattering from non-spherical particles in the marine atmosphere. As the modeling is refined, greater insights into the nature of the contents of the marine atmosphere should be achieved.

IMPACTS/APPLICATIONS

Because the marine atmospheric boundary layer aerosols, and therefore their polarization properties, are highly variable, it is important to understand that variation in order to optimize polarimetric sensing and imaging systems. By developing techniques to rapidly quantify the optical properties of aerosols in the MABL, it will be possible to make measurements to provide a more quantitative understanding of the effect of polarization on imaging techniques. Measurements will establish the significance of the nature of the aerosols to visibility and image enhancement.

TRANSITIONS

Scanning polarization-modulation nephelometry is presently being developed for use to measure the particulate matter in diesel engines and to determine fiber orientation in the paper industry.

RELATED PROJECTS

We have continued the collaboration with Prof. Patricia Hull of Tennessee State University. During her summer spent in Berkeley and she developed an alternative method of fitting the data based on neural nets that looks very promising to improve the speed and accuracy of the fitting algorithms. We are presently developing a Diesel Particle Scatterometer (DPS) dedicated to the measurement of diesel exhaust. The DPS uses the same polarization modulation and phase sensitive detection used in bistatic nephelometer but is specialized for rapid *in situ* measurements of diesel exhaust stream. In the scatterometer the angle dependence is measured nearly simultaneously using a multiplicity of fixed detectors. We have also used polarization nephelometry to determine fiber orientation in paper webs for the fiber industry.

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